Combining organic-rich coacervates with hydrothermal vent systems as a model for prebiotic compartmentalization

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Protocells can be defined as micro-systems that have aspects of basic metabolism, replication or compartmentalization of biomolecules. Coacervate is one of the several protocell models suggested in the origin of life scenarios. Prebiotic organic macromolecules could have likely undergone liquid-liquid phase separation, even at very low concentration resulting in organic-rich phase, or "coacervates". Organic-rich coacervates can accumulate biomolecules inside and have lower water content. This would have been beneficial in prebiotic condition, in which biomolecules are expected to be dilute. However, characterization of coacervates in prebiotic relevant conditions has not been explored yet. Therefore, my research aims to evaluate coacervate droplets as potential prebiotic compartments in various geological conditions.

Hydrothermal vents have gained significant interest as a prebiotic geological site, due to its catalytic mineral surface, self-organized pore structures and temperature/redox gradient. Recent findings from

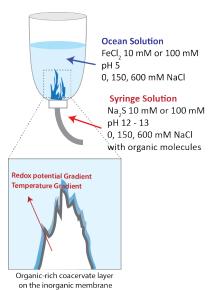


Figure 1. Illustration of laboratory hydrothermal chimney setup with organic molecules composing coacervates.

of hydrothermal chimneys

formed by the injection methods

NASA missions suggest the possibility of emergence on life in the ocean or hydrothermal vents of early Earth, Europa and Encladus. Mineral surfaces, as well as redox gradients by hydrothermal vents could aid the coacervate formation. Hence, I proposed to spend three weeks at the NASA Jet Propulsion Laboratory (JPL) collaborating with Dr. Laurie Barge to learn about laboratory hydrothermal vent systems. With my knowledge on coacervate formation by LLPS and Dr. Barge's expertise in laboratory hydrothermal systems, I tested the coexistence of our coacervate systems with hydrothermal chimney systems.

During my visiting at NASA JPL, I investigated the ironsulfide (FeS) chimney formation under various salt, ferrous and sulphide concentration with/without organic molecules (Figure 1). Prebiotic ocean is expected to have 600 mM of salt, 10 mM Fe²⁺ and 10 mM S²⁻ approximately. Various salt concentration did not affect the chimney morphology in terms of chimney or mineral mound formation. The concentration of ferrous ion and sulfide ion was more critical on chimney formation: 100 mM of each FeCl₂ and Na₂S tend to form hydrothermal chimney structure (Figure 2 A), whereas 10 mM of each

FeCl₂ and Na₂S did not form chimney morphology but mineral is suspended in ocean solution (Figure 2 B). Therefore, we proceeded to test 10 mM of FeCl₂ and Na₂S to check more noticeable impact of charged organic molecules on hydrothermal chimney formation.

Coacervates composed of nucleotide (adenosine triphosphate) (ATP) with poly (allyl amine) (PAH) was chosen to mimic coacervates; because i) these molecules are costly less expensive, ii) coacervate formation can be occurred spontaneously in pH 5 - 10 and up to 600 mM NaCl solution, and iii) this

coacervate has been characterized well in literature. Interestingly, injection of PAH and ATP aided to form the hydrothermal chimney-like morphology with solid support (Figure 2 C), whereas the system without coacervate molecules did not have chimney- or mound-like morphology (Figure 2 B). Visually noticeable white layer was formed by injection of organic molecule layer as seen in Figure 2 D. Charged organic molecules itself or coacervate layer at the FeS mineral surface would have provided attractive intermolecular forces that might have led this morphology. I am characterizing this system to investigate the wetting of coacervate droplets on FeS minerals using optical microscopy and Raman microscopy at Penn State. We will develop microfluidic devices for hydrothermal vent systems with organic molecules to validate whether this white layer can be coacervates, and furthermore behave as protocells. These preliminary results suggest the promising future collaboration with novelty in the first demonstration of coexistence of coacervates under hydrothermal vent conditions and exploration of potential roles for these structures in emergence of life.

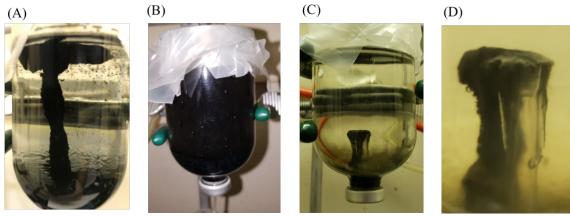


Figure 2. Laboratory FeS hydrothermal vent systems in various conditions. (A) 100 mM FeCl₂, 100 mM Na₂S without NaCl. (B) 10 mM FeCl₂, 10 mM Na₂S with 600 mM NaCl. (C) 10 mM FeCl₂, 10 mM Na₂S with 600 mM NaCl with PAH (190 μ M, 50 mM in charge units) and ATP (12.5 mM, 50 mM in charge units). (D) Magnified image of (C).

From this travel supported by the NAI funding, I was able to build new professional connections with JPL that would propel future collaborations and proposals in astrobiology field. I had weekly meetings and other individual meetings with scientists and engineers at NASA JPL, which was great opportunity to interact actively with scientists/engineers in astrobiology across disciplinary boundaries and broaden my perspectives in astrobiology and NASA missions. The NAI funding has helped not only to develop my research and career goals, but also to embark the collaboration with Dr. Barge at JPL. We are excited by these preliminary results and looking forward to continue this collaboration for further characterization and understanding of hydrothermal vents with coacervates.